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GENERATION OF COHERENT VUV AND SOFT X-RAYS.

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I. INTRODUCTION

The goal of this program is the development of practical sources of coherent vacuum ultraviolet radiation. During this reporting period two primary projects have been active. The first is the production of coherent radiation at the Lyman α wavelength of 1218 Å. Such radiation is potentially applicable to the diagnosis of plasmas, such as those studied under the nuclear fusion program. The second project has elucidated the factors which limit the efficiency of the process 3547 Å $\rightarrow$ 1182 Å in Xe, and suggests solutions. Details of the research are presented in the following sections.

II. GENERATION OF COHERENT RADIATION AT LYMAN α WAVELENGTH

(K. S. Hsu, J. F. Young, and S. E. Harris)

The goal of this program is the development of practical sources of coherent extreme ultraviolet and soft x-ray radiation. During the past six months we have worked on the generation of coherent, short pulse length, Lyman α (1216.7 Å) radiation; such radiation is one of the major diagnostic tools for the thermal nuclear fusion program. The general method employed is optical harmonic generation in gaseous xenon. We have investigated two different approaches to reach this goal. The first is to combine two photons of 2660 Å and a single photon of 1.4085 μm in Xe, taking advantage of resonance enhancement to create a large nonlinear coefficient. The second approach is direct third harmonic generation of 3650 Å in Xe. Our results are described in the following sections.

A. Frequency Mixing Approach (2 × 2660 Å + 1.4085 μm)

The object of this experiment is to enhance the efficient of a three-photon sum process in Xe. While a single photon of 2660 Å is about 30,000 cm^{-1} below the $6s[3/2]_1^0$ state, two photons of 2660 Å is only 2,961 cm^{-1} below the $6p[5/2]_2$ level (a two-photon allowed transition state), and 1216.7 Å is only 2,272 cm^{-1} above the $5d[1/2]_1^0$ state.[1] Thus the dominant perturbation path is:

$$5p^6\ {}^1S_0 \rightarrow 6s[3/2]_1^0 \rightarrow 6p[5/2]_2 \rightarrow 5d[3/2]_1^0 \rightarrow 5p^6\ {}^1S_0$$

The relevant energy level diagram is shown in Fig. 1. Though not all of the dipole matrix elements are available, estimation of the third-order nonlinear susceptibility is possible because this process has the same dominant path as tripling 3547 Å to generate 1182 Å in Xe. The two nonlinear susceptibilities differ only in their detunings. Our calculation shows that the $\chi^{(3)}$ for this proposed process should be 3.19×10^{-34} esu, or 5.7 times larger than that for the process of tripling 3547 Å to generate 1182 Å.[2]

The index of refraction of Ar, Kr, and Xe at the Lyman α line has been measured by Gill and Heddle using the Rayleigh scattering methid, as[3]

$$n - 1 = 5.65 \times 10^{-4} \quad \text{for Ar}$$

$$n - 1 = -53.1 \times 10^{-4} \quad \text{for Kr}$$

$$n - 1 = -172.1 \times 10^{-4} \quad \text{for Xe}$$

Although the measurements of Kr and Xe were considered to be too high because the absorption band of Xe_2 and Kr_2 molecules are located in the vicinity of the resonance lines of Xe and Kr and thus distorted the measurement of the refractive indices, it was almost certain, according to Gill and Heddle, that the $n - 1$ values for Kr and Xe were negative.[3-6] Based on the construction of the Sellmeier equation by the measured refractive indices for wavelengths longer than 1680 Å, Chashchina and Shreider predicted indices of refraction at the Lyman α line as

$$n - 1 = 2.95 \times 10^{-5} \quad \text{for Kr}$$

$$n - 1 = 8.88 \times 10^{-4} \quad \text{for Xe}$$

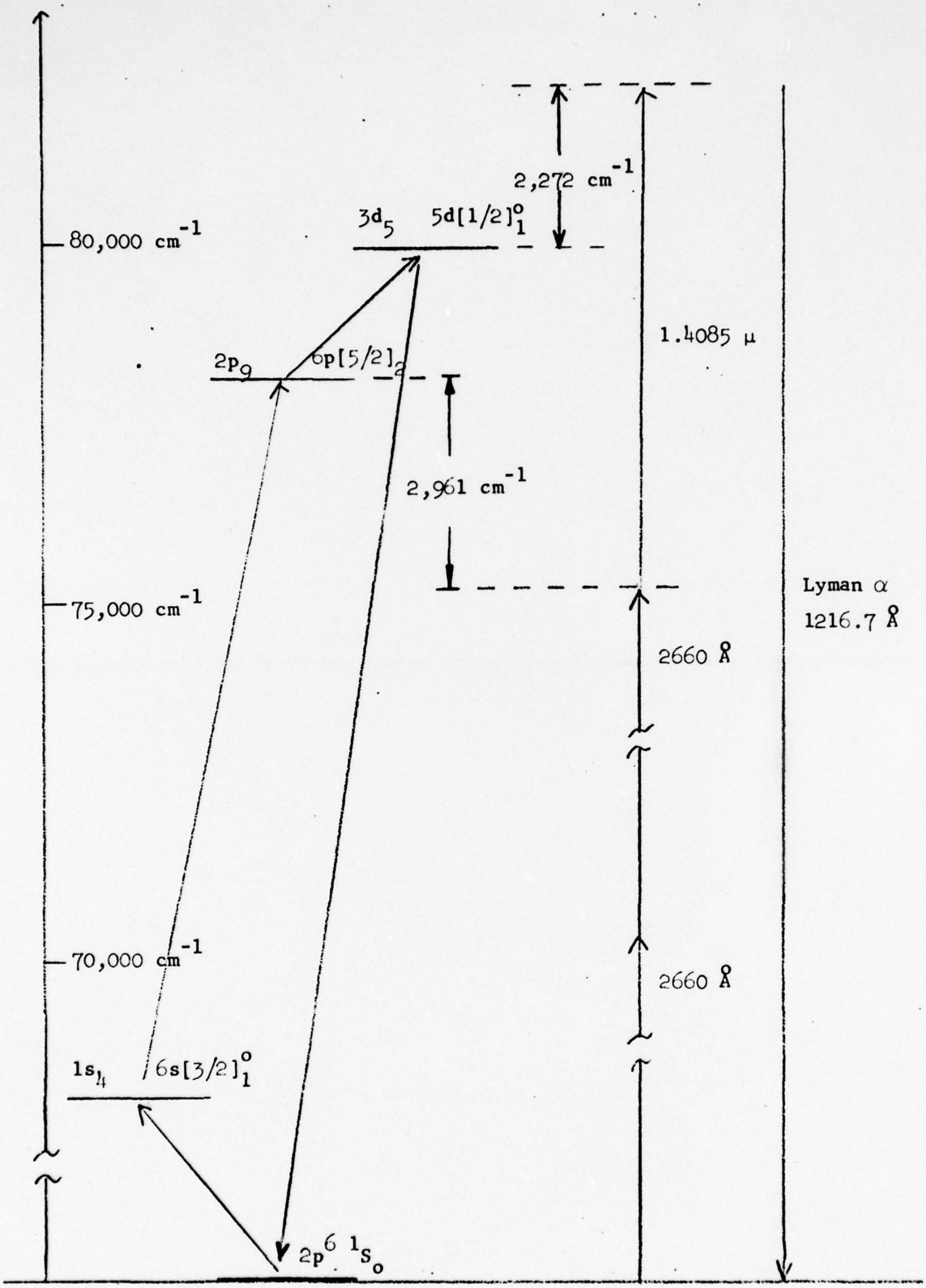

Fig. 1--Energy level diagram of XeI.

The indices of refraction of Xe at 2660 Å and 1.4085 μm have been precisely measured to within an 0.1% error as[7]

$$n - 1 = 7.99 \times 10^{-4} \quad \text{for 2660 Å in Xe}$$

$$n - 1 = 6.78 \times 10^{-4} \quad \text{for 1.4085 μm in Xe}$$

Using the data of Gill and Heddle,[3] the wave vector mismatch ($\Delta k \equiv k_{sum} - k_1 - k_2 - k_3$) of the proposed process is $- 9.36 \times 10^3$ cm^{-1} for standard temperature and pressure conditions (i.e., 273°K and 1 atm), compared to - 18.3 cm^{-1} using the estimation of Chashchina and Shreider. Thus, by both measurements, the process should be negatively dispersive and therefore phasematchable using a positive dispersive gas.[8,9] The power conversion efficiency from 1.4085 μm to 1216.7 Å is given by

$$\frac{P_{(1216.7\ \text{Å})}}{P_{(1.4\ \mu m)}} = 1.48 \times 10^{-54}\ N^2 \chi^{(3)^2} P^2_{(2660\ \text{Å})} |I|^2 \quad \text{mks}$$

where N is the Xe number density and $|I|^2$ is the focusing integral of Ward and New which has a maximum value of 5.34 at $b\Delta k = - 2$ for the third-order process under the tight focusing condition.[10,11] The power density of the input radiation is limited by the gas breakdown of Xe and was estimated as about 5×10^{12} W/cm^2.[12] With 200 MW of 2660 Å radiation confocally focused to 6 cm, 14 torr of Xe will satisfy the optimized focusing condition. The power conversion efficiency from 1.4085 μm to 1216.7 Å can be more than 10%. For 14 torr of Xe, the number density of Xe_2 molecules is 5.19×10^{13}; the absorption cross section of Xe_2 at the Lyman α line was measured to be 1.87×10^{-17} cm^2.[5] This yields an absorption depth of about 10^3 cm; thus Xe_2 absorption should not be a problem.

The experimental set-up is shown in Fig. 2. We have tried direct single-pass pumping of a $LiNbO_3$ parametric generator followed by a $LiNbO_3$ parametric amplifier to generate 1.4085 μm. We found that this scheme is highly unstable and unable to provide a good spatial mode because of the extremely high gain of the parametric crystal. We are investigating the possibility of using a synchronously matched cavity scheme pumped by a passively mode-locked Nd:YAG laser pulse train, similar to the matched cavity pumped dye laser devised by Goldberg and Moore.[13]

In summary, this frequency mixing approach takes advantage of resonant enhancement of the third-order nonlinearity and the availability of high power 2660 Å radiation. The major difficulties are the generation of 1.4085 μm and the attainment of good beam overlap, both spatially and temporally. Further development of the 1.4085 μm source is currently underway.

B. <u>Direct Tripling Approach (3 × 3650 Å)</u>

The object of this experiment is to generate 1216.7 Å by directly tripling 3650 Å in Xe to avoid the experimental difficulty of beam overlap encountered in the frequency mixing approach described in Section A. Calculations show that the nonlinear susceptibility and medium dispersion are quite comparable to those of the 3547 Å direct tripling process. Thus 50 μJ of 3650 Å in 20 ps will be sufficient to generate a detectable 1216.7 Å signal. We plan to use the matched cavity scheme devised by Goldberg and Moore[13] to generate 3650 Å by pumping BDBP dye with a pulse train of 3547 Å; the experimental set-up is shown in Fig. 3. Construction of the dye laser is currently underway.

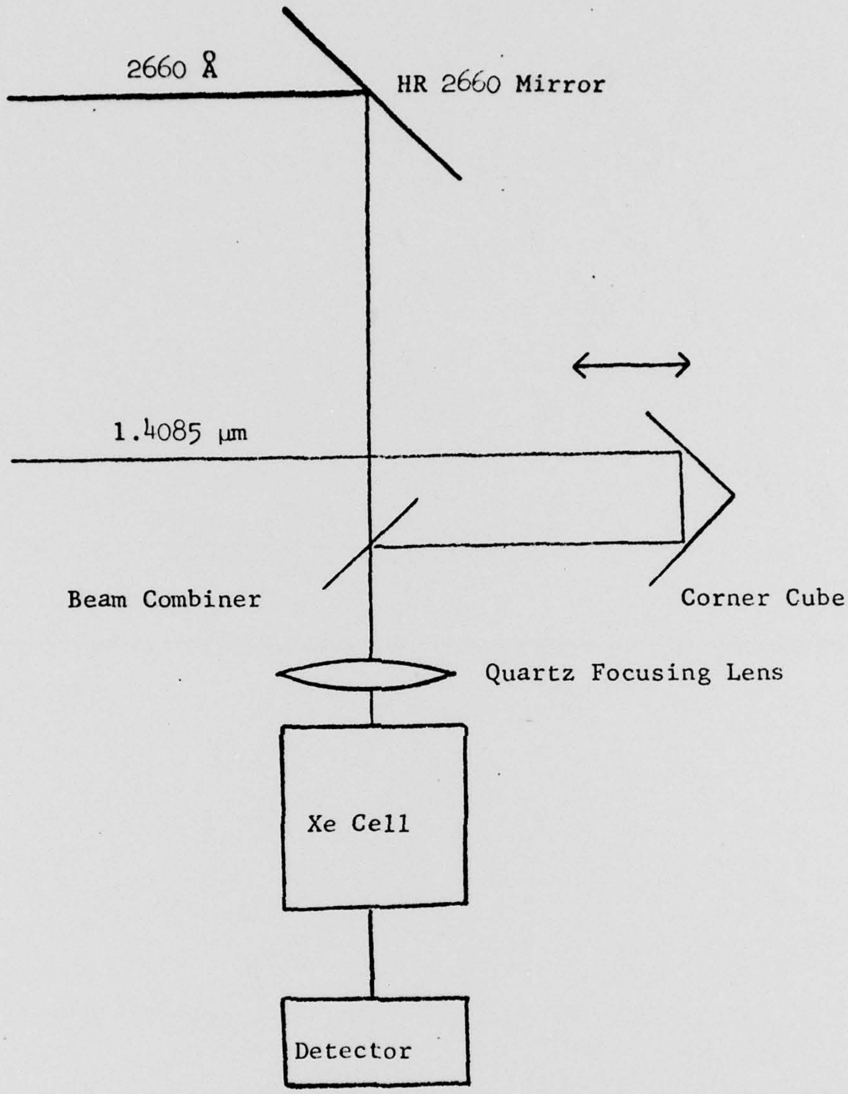

Fig. 2--Experimental set-up for the mixing frequency approach.

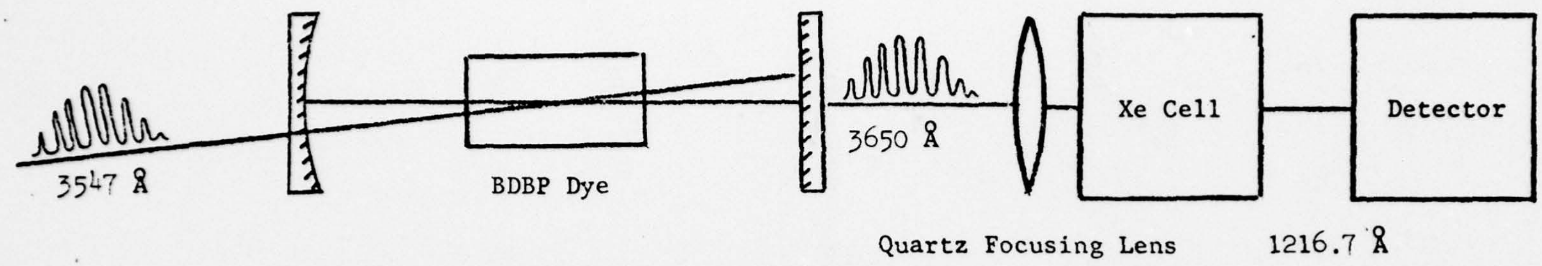

Fig. 3--Experimental set-up for the direct tripling approach.

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III. LIMITATION OF 1182 Å GENERATION EFFICIENCY IN Xe

(L. J. Zych, J. F. Young, and S. E. Harris)

The recent demonstration of large gains in discharge-pumped XeF for single 30 ps 3547 Å pulses[1] indicates that very high power, high energy sources of 3547 Å radiation should be practical. This fact has motivated us to explore the factors which limit the ultimate efficiency of the tripling process 3547 Å → 1182 Å in Xe phasematched with Ar, as first demonstrated by Kung, Young, and Harris.[2]

The nonlinear susceptibility measured in Ref. 2 indicated that for reasonable experimental parameters, conversion efficiencies of several percent should be feasible. Clearly, such efficiencies coupled with amplified high power 3547 Å pulses could lead to a powerful source of 1182 Å radiation for microlithography, holographic microscopy,[3] and for the generation of extremely short wavelengths using additional nonlinear processes.[4] However, despite the work of Ref. 2 and subsequent experiments in our laboratory, observed efficiencies have reached only about 0.1%. The cause of the low efficiencies has been attributed to loss, poor gas mixing, and bad laser mode quality. During the past six months we have completed a series of measurements which indicate that the observed efficiencies are in fact limited by the quadratic Kerr effect. This is a third-order process in which large power densities at 3547 Å can produce changes in the index of refraction at <u>both</u> 3547 Å and 1182 Å, thus effecting phasematching. Careful absorption

measurements indicated that neither Xe, Xe_2, nor normal impurity absorption is present under practical cell lengths and pressures. In addition, a premixing method was used to insure gas mix homogeneity.

The THG efficiency can be expressed as[5]

$$\mathcal{E} \propto N^2 \chi^2 P^2 \cdot F_1(b\Delta k) \tag{1}$$

where P is the power at 3547 Å and $F_1(b\Delta k)$ depends on the focusing and medium dispersion. The k vector mismatch in the presence of the Kerr effect is

$$\begin{aligned} \Delta k &= k(3\omega) - 3k(\omega) = \frac{6\pi}{\lambda_1}[n(3\omega) - n(\omega)] \\ &= \frac{6\pi}{\lambda}[\Delta n_0 + \Delta n_k] \cdot N \end{aligned} \tag{2}$$

where Δn_0 is the zero-field index mismatch per atom, and Δn_k is the per atom index mismatch due to the Kerr effect:

$$\begin{aligned} \Delta n_k &= 1.1 \times 10^{13} \, [\chi_k(3\omega) - \chi_k(\omega)] \, (P/A) \quad \text{mks} \\ &= 0.013 \, [\chi_k(3\omega) - \chi_k(\omega)] \, (P/A) \quad \text{esu} \\ &\equiv \beta \cdot P/A \end{aligned} \tag{3}$$

Note that the value of Δn_0 can be adjusted by varying the Xe:Ar ratio.

In our experiments the conversion efficiency was measured at various values of power density as N was varied. As shown in Ref. 5, the efficiency

has a maximum at a value of $b\Delta k = -4$, which corresponds to a particular N. We can use Eq. (2) in Eq. (1) to find these peak (phasematched) efficiencies

$$\mathcal{E}_p = 6.9 \times 10^{-4} \cdot \chi^2 \cdot \frac{(P/A)^2}{(\Delta n_0 + \beta P/A)^2} \quad \text{(esu)} \tag{4}$$

when the number density is adjusted so that

$$(b\Delta k) = -4 \quad \text{or}$$

$$(\Delta n_0 + \beta P/A) = -4\lambda_1/6\pi b N \tag{5}$$

Figure 1 shows measured values of $\mathcal{E}_p$ as a function of P/A for pure Xe, while Fig. 2 shows the pressure of Xe at which $\mathcal{E}_p$ occurred. The focusing conditions were held constant: a confocal parameter of $b = 1$ cm located in the center of a 50 cm cell. For low values of P/A the optimum Xe pressure is constant, and from Fig. 2 and Eq. (5)

$$\Delta n_0 = -3.3 \times 10^{-23} \text{ cm}^3 \quad \text{or}$$

$$NL_c = 1.8 \times 10^{17} \text{ cm}^{-2} \tag{6}$$

Using Eq. (4) we find

$$\chi = 2.3 \times 10^{-35} \text{ esu} \tag{7}$$

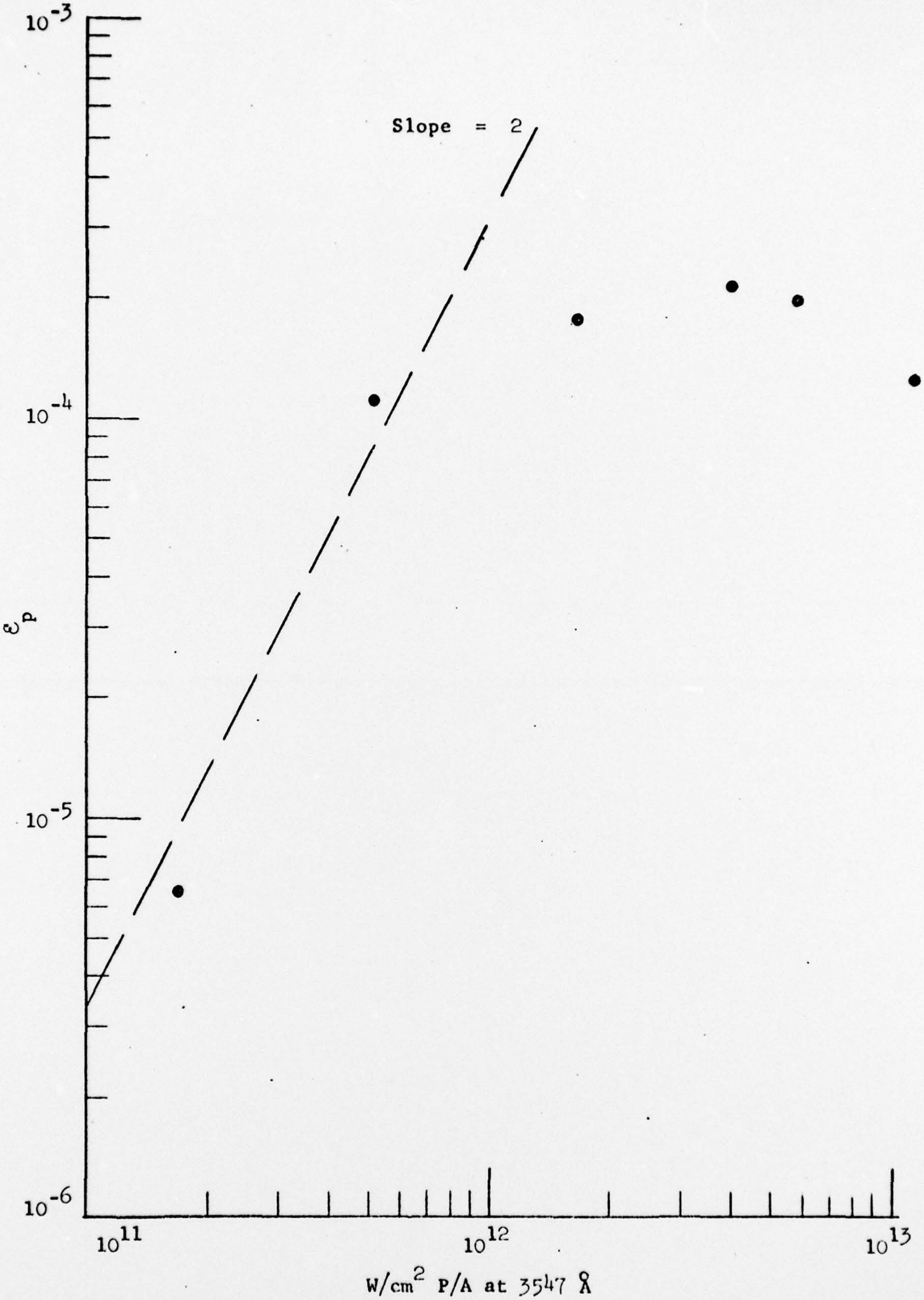

Fig. 1--Measured peak conversion efficiency for 3547 Å → 1182 Å vs. P/A in pure Xe.

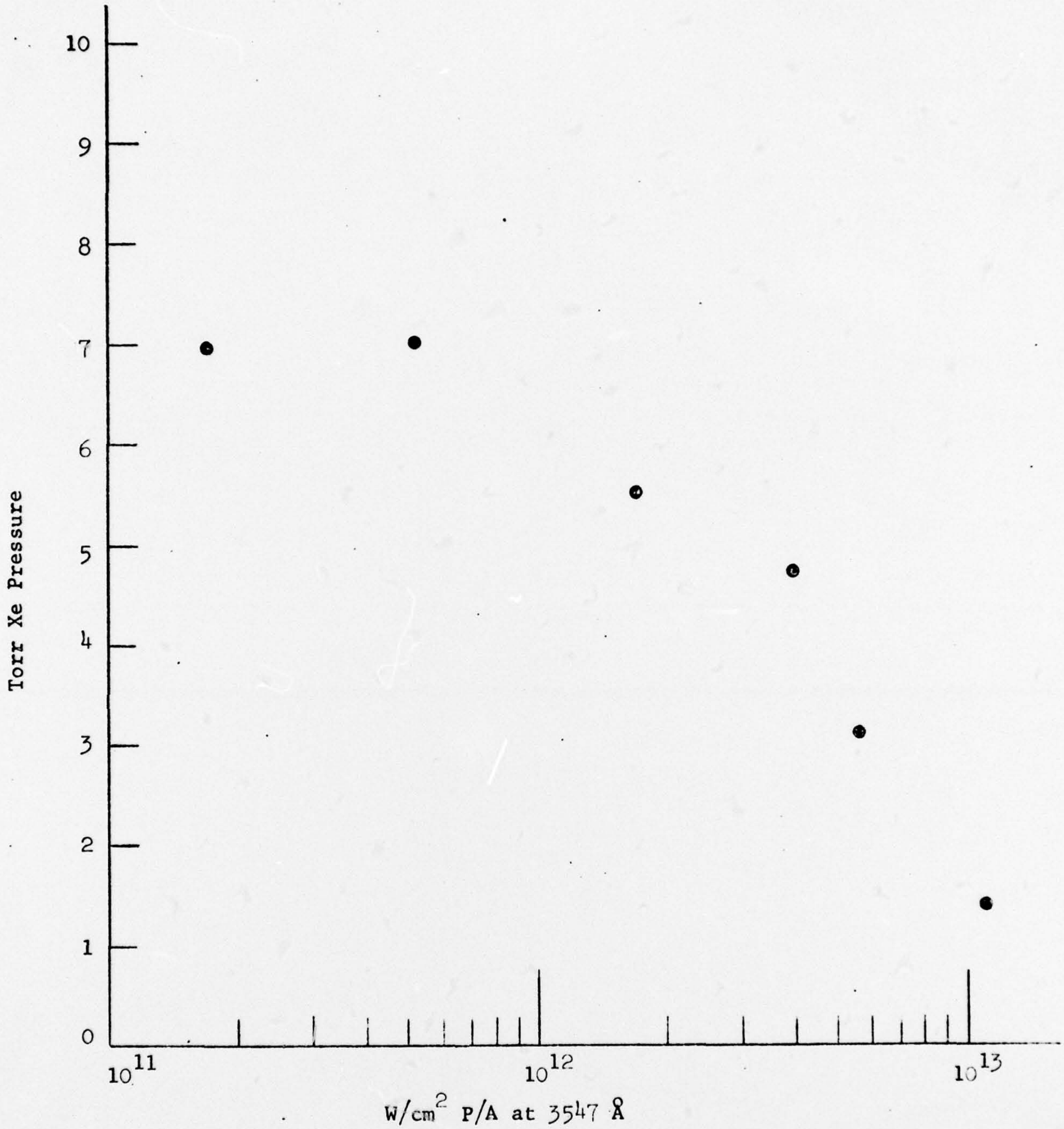

Fig. 2--Pressure of pure Xe for maximum conversion efficiency as a function of P/A .

At higher P/A , lower Xe pressures are required to optimize $\mathcal{E}$; this, along with Eq. (6), implies that $\beta < 0$. In Fig. 2 the optimum Xe pressure drops from 7 torr at low P/A to 3.5 torr at $P/A = 5.2 \times 10^{12}$ W/cm^2 , and this implies

$$[\chi_k(3\omega) - \chi_k(\omega)] = -4.9 \times 10^{-34} \quad \text{esu} \tag{8}$$

For large incident power densities such that

$$|\beta P/A| \gg |\Delta n_0| \tag{9}$$

the Kerr effect will dominate the dispersion of the medium and the efficiency will be limited to a maximum value of

$$\mathcal{E}_{max} = 4.1 \left[\frac{\chi}{\chi_k(3\omega) - \chi_k(\omega)} \right]^2 \tag{10}$$

This limit is independent of all other experimental parameters, including *the Xe:Ar ratio* (Δn_0) *, so* long as Eq. (9) is satisfied. For operation well into the Kerr regime however, $\mathcal{E}_{max}$ can only be achieved using an input pulse of constant P/A . Although this analysis has been done for the tight focusing case, Eq. (10) is also valid for the plane wave case. From Eqs. (7), (8), and (10) we estimate that for Xe

$$\mathcal{E}_{max} = 9 \times 10^{-3} \tag{11}$$

This indicates that the limiting efficiency of 2×10^{-4} in Fig. 1 is not a Kerr limitation, but rather due to some other effect which becomes

important at large P/A, such as multiphoton absorption or breakdown. To circumvent this problem we shifted the Kerr dominated region to lower P/A by reducing Δn_0 using a 1:11 Xe:Ar mixture. The gases were measured into a cylinder and allowed to mix thoroughly for a day or more, to insure homogeneity of the test gas. With this mixture efficiencies as high as 2×10^{-3} were observed, again limiting at about 5×10^{12} W/cm^2. Further reduction of Δn_0 should result in even closer agreement with Eq. (11).

Theoretical estimates of the Kerr susceptibilities in Xe indicate that the dispersion is probably dominated by the $\chi_k(3\omega)$ term, specifically $\chi(3\omega, \omega, -\omega)$. It appears that a major contribution to the susceptibility results from the interaction of the 5d Xe level and the autoionization states.

These results indicate that the ultimate conversion efficiency in the Xe:Ar system is limited to less than 1% by the Kerr effect. It may be possible to overcome this limitation by adding an additional atomic species with a compensating Kerr susceptibility; however, the species used must not interfere radically with Δn_0 or χ. Mg appears to be a good candidate. It is negatively dispersive, has a substantial χ for 3547 Å → 1182 Å generation,[6] and we estimate a $\chi_k(\omega) \approx -4.6 \times 10^{-33}$ esu. Thus a ratio of 9.4:1 of Mg:Xe should eliminate the Kerr effect; clearly, complete, uniform cancellation in Eq. (10) is not required to achieve significant efficiencies.

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IV. FUTURE EFFORT

Future effort on the Lyman α generation project was presented in Section II. Experimental work on the generation of 1182 Å radiation has been suspended pending further calculations, and in order to commence work on a new project: development of a high brightness, high resolution, picosecond time scale VUV light source based on spontaneous anti-Stokes scattering.

V. PUBLICATIONS

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